1 Rainfall redistribution in subtropical Chinese forests changes over 22

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17 Abstract

Rainfall redistribution through the vegetation canopy plays a key role in the hydrological cycle. Although there have been studies on the heterogeneous patterns of rainfall redistribution in some ecosystems, the understanding of this process in different stages of forest succession remains insufficient. Therefore, this study investigated the change tendency of rainfall redistribution and rainwater chemistry in a subtropical forest succession in South China, based on 22 years (2001–2022) of monitoring rainfall (740 valid events).

25 Results showed that at the event scale both throughfall ratio and stemflow ratio in pine forest (PF) were higher than in mixed forest (MF) and broadleaf forest (BF). At 26 the interannual scale, throughfall and stemflow of forests experienced an initial 27 decrease followed by a subsequent increase over the entire measurement period (except 28 stemflow of the pine forest), which reflects the trend of open rainfall. The variability of 29 throughfall showed an increase from MF to PF to BF, and the variability of stemflow 30 likewise showed an increase from MF to PF to BF. Changes of throughfall and stemflow 31 32 in the broadleaf forest are thus higher than those in the mixed forest and pine forest over time. Furthermore, important differences in rainwater chemistry fluxes among the 33 three forest types were found, changing in varying order over time. On average, total 34 nitrogen (TN) and total phosphorus (TP) fluxes of throughfall increased from BF to MF 35 to PF, while the potassium (K^+) flux of throughfall showed a decrease from BF to MF 36 to PF. Stemflow chemical fluxes varied less among forest types and over time, though 37 38 tree species most importantly affected varying stemflow chemistry.

These results show important changes in patterns of rainfall redistribution over time and that characteristic variations are driven by rainfall and forest factors. This study thus provides insight into long-term rainfall redistribution processes by linking changes in rainfall spectra with a typical subtropical forest succession sequence.

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44 Keywords: throughfall, stemflow, forest types, forest meteorology, long-term study

45 **1 Introduction**

In recent years, there has been on-going concern about the potential impacts of 46 climate change on forest ecosystems, particularly regarding rainfall input to water 47 resources (Reynaert et al., 2020; Grossiord et al., 2017; Bruijnzeel et al., 2011; 48 Leuzinger and Körner, 2010). Numerous studies have documented rainfall regimes and 49 their effect on the water cycle in different regions of the world, including spatial and 50 temporal changes in the amount, intensity, and frequency (Brasil et al., 2018; Ponette-51 González et al., 2010). Meanwhile, these variables in rainfall refer to the redistribution 52 53 of rainfall into canopy interception, throughfall and stemflow, being important components of hydrological processes in terrestrial ecosystems (Germer et al., 2010; 54 Levia and Frost, 2006; Loustau et al., 1992). Rainfall redistribution patterns can impact 55 biogeochemical cycles down to soil moisture distribution, which in turn affects the 56 activity of soil microorganisms that decompose organic matter (Tonello et al., 2021a; 57 Junior et al., 2017; Van Stan II and Pypker, 2015). For example, Sun et al. (2023) 58 59 showed that throughfall reduction significantly affects the soil carbon cycle in a 60 subtropical forest. Therefore, understanding the roles of rainfall redistribution in woodlands is essential. 61

62 Rainfall redistribution, as the partitioning into interception loss, throughfall and stemflow, is an important hydrological process that regulates water and nutrient cycling 63 in forest ecosystems. Interception loss refers to the part of the event rainfall intercepted 64 by the canopy, accounting for about 10%-30% of gross rainfall depending on the 65 studied forest canopy, e.g. shrub (Zhang et al., 2015), mixed broadleaf (Yan et al., 2003), 66 or pine (Loustau et al., 1992). The remaining rainwater reaches the ground either as 67 throughfall or stemflow. Throughfall is a critical component of rainfall redistribution, 68 and it contributes on average to approximately 60%-90% of the gross rainfall on the 69 70 floor in forests, shrubland, or cropland. (Zhang et al., 2023; Zhang et al., 2021; Brauman et al., 2010; Marin et al., 2000). Raindrops coalesce or splash on canopy leaf surfaces, 71 generating spatially different throughfall volume and raindrop kinetic energies which 72 can be larger or lower than that of open rainfall (Levia et al., 2019; Goebes et al., 2015). 73 74 Stemflow, the water flowing bottomwards along the plant stem or trunk, often accounts for only a small proportion (0–12%) of rainfall (Niu et al., 2023; Yue et al, 2021; Llorens 75 and Domingo 2007). Nevertheless, stemflow inputs can be important as hot spots for 76 near-trunk soils, inducing water and nutrient enrichment and deep infiltration, but also 77 erosion (Zhao et al., 2023; Llorens et al., 2022). It can funnel more water than open 78

rainfall on an equivalent area and contributes to 10% of the annual soil water input
(Levia and Germer, 2015; Chang and Matzner, 2000). Throughfall and stemflow restrict
water input to the soil layer, thereby affecting soil moisture conditions, runoff
generation and water and nutrient cycling (Lian et al., 2022; Lacombe et al., 2018; Klos
et al., 2014).

84 The proportions of rainfall redistribution are generally driven by meteorological conditions (e.g., rainfall amount, intensity, duration) and vegetation cover (e.g., canopy 85 structures, tree characteristics) (Tonello et al., 2021a; Sun et al., 2018; Mużyło et al., 86 87 2012; Nanko et al., 2006). For meteorological conditions, numerous studies have documented that throughfall volume and stemflow volume increase with increasing 88 gross rainfall and intensity (Ji et al., 2023; André et al., 2011). The ratios of throughfall 89 and stemflow were both characterized as logarithmically increasing with rainfall, 90 tending to be quasi-constant for heavy rainfall events (Zhang et al., 2021; Liu et al., 91 92 2019). This is also synchronously related to the gradual saturation of the canopy which limits the ratios of rainwater partitioning (Carlyle-Moses et al., 2004). Besides, 93 94 differences in water volume spatially exist from place to place. The spatial variability (expressed as a coefficient of variation) of throughfall volume is generally higher for 95 96 small rainfall events (< 10 mm) than for heavy rainfall events (Germer et al., 2006; Price et al., 1997). 97

98 Rainfall redistribution among different plant species can vary significantly due to 99 differences in the structure and characteristics of their canopies. Specifically, key 100 factors determining the redistribution of rainfall such as leaf area index (LAI), leaf 101 shapes and orientations can affect the amount of intercepted loss and throughfall (Zhang 102 et al., 2021; Goebes et al., 2015; Keim et al., 2006). The diameter at breast height (DBH), 103 bark type and orientation of trunks/stems and branches influence the amount of 104 stemflow (Levia et al., 2015; Livesley et al., 2014; Germer et al., 2010). For each of the rainfall partitioning fluxes, their responses to the influential predictors often show high 105 variation. A modelling study of rainfall partitioning in China explained that throughfall 106 was best represented by mean tilt angle (MTA), followed by DBH. Subsequently, DBH 107 108 was the dominant predictor for stemflow, followed by LAI and bark texture (Zhang et al., 2023). Due to these factors, rainfall redistribution presented different degrees of 109 spatial variability. This variability (expressed as coefficient of variation) decreased with 110 increasing rainfall amount and intensity, consequently tending to be quasi-constant 111 (Germer et al., 2006). Besides, at interannual scale, ratios of rainfall redistribution are 112

driven by annual canopy structures. The study of Niu et al. (2023) documented that the annual throughfall ratio gradually increased, while the annual stemflow ratio and interception loss ratio decreased with increasing thinning intensity in shrub plantations. Meanwhile, annual changes of rainfall events (amount and intensity) reinforced the time instability of throughfall spatial variability (Rodrigues et al., 2022). Overall, the rainfall-canopy interactions play a key role in rainfall redistribution processes and further affect the water cycle in many ecosystems.

120 The vegetation canopy is the functional interface between an ecosystem and 121 atmospheric wet deposition (Van Stan II and Pypker, 2015). Leaves and trunks/stems, acting as a filter, alter rainwater chemical concentrations via leaching and depositing 122 processes. As a result, throughfall and stemflow exhibit high chemical concentrations 123 compared to open rainfall (Jiang et al., 2021; Zimmermann et al., 2007). For instance, 124 in a Chinese pine plantation, the volume-weighted mean concentrations of NH4⁺ and NO3⁻ 125 in throughfall were significantly higher than those in open rainfall (Wang et al., 2023). 126 127 Stemflow ion fluxes (e.g., K⁺) from deciduous tree species were higher than those for evergreen tree species because of the differences in bark morphology and branch 128 architecture (Su et al., 2019). Moreover, it is also common that throughfall and 129 stemflow chemistry show fluctuating seasonality with the shifts in rainfall regime and 130 131 leaf growth (Turpault et al., 2021; Siegert and Levia, 2014; Staelens et al., 2007). Plants mainly require N, P, K, Ca and Mg. In general, N, K and Ca are the most important 132 133 inputs to a forest ecosystem, and P is the least. Phosphorus (P) is considered to be a limiting nutrient element in tropical and subtropical forests. The long-term productivity 134 of vegetation depends on the input of atmospheric P. Besides, an increasing trend of 135 atmospheric nitrogen (N) deposition with more frequent seasonal droughts in 136 subtropical areas of China was reported (Zhou et al., 2011), and may inhibit the growth 137 138 of plants and affect the productivity and functioning of forest ecosystems (Wu et al., 2023; Borghetti et al., 2017). Therefore, the important significance of atmospheric 139 precipitation to an ecosystem can also be confirmed for element cycling, and canopy 140 141 leaching plays an important role in the chemical cycle of forest ecosystems.

Although there have been studies on the spatiotemporal variability of rainfall redistribution, most of these are limited to data from short-term monitoring over several months to one-two years (Liu et al., 2019; Ziegler et al., 2009; Carlyle-Moses, 2004; Marin et al., 2000). There are few studies exceeding several years of measurements and simultaneously focusing on forest structural changes and rainwater interception (Grunicke et al., 2020; Shinohara et al., 2015; Jackson, 2000). Long-term field
monitoring studies are valuable to gain insight into the temporal dynamics of forest
hydrological processes (Rodrigues et al., 2022; Sun et al., 2023; Levia and Frost, 2006).
Such studies can also contribute to identifying patterns and trends in rainfall
redistribution, which is essential for predicting the long-term effect of water resource
change on forest ecosystems.

Therefore, in this study, we focus on the changing characteristics of throughfall 153 and stemflow in a subtropical forest succession sequence (pine forest -> mixed forest -> 154 155 monsoon evergreen broadleaf forest), based on long-term monitoring. Specifically, the objectives are to analyze: (1) the changes in water volume of throughfall and stemflow, 156 157 and (2) the changes in water chemistry (N, P, K^+) of rainfall, throughfall and stemflow among the three forests. We hypothesize that: (1) both throughfall and stemflow in the 158 broadleaf forest are characterized by high variability compared to mixed forest and 159 followed by pine forest, and (2) the chemical fluxes of throughfall and stemflow change 160 over time from broadleaf forest to mixed forest to pine forest. We aim to assess the 161 variability of forest hydrological processes from a long-term perspective to help predict 162 future dynamic trends of water resources in subtropical forest ecosystems. 163

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165 2 Materials and methods

166 **2.1 Study site**

This study was conducted at the Dinghushan Biosphere Reserve ($23^{\circ}09' 21'' N \sim$ 167 23°11′ 30″ N, 112°30′ 39″ E ~ 112°33′ 41″ E) located in Zhaoqing City, South China. 168 169 Dinghushan catchment consists of two streams both with 12 km length, which flow into the West River (the main trunk of the Pearl River). According to the Köppen-Geiger 170 climate classification (Kottek et al., 2006), the study area belongs to tropical monsoon 171 climate (Cwa) with pronounced wet (April-September) and dry season (October-172 March). The average annual temperature is 20.9 °C, and the annual rainfall and 173 evaporation are 1900 mm and 1115 mm, respectively. Dinghushan Biosphere Reserve 174 is covered with a complete horizontal succession series of three types of subtropical 175 forest, which is highly representative of the region (Zhou et al., 2011). Monsoon 176 evergreen broadleaf forest (BF) is 400 years old with typical tree species including 177 Castanopsis chinensis (Spreng.) Hance, Schima superba Gardner & Champ., 178 179 Cryptocarya concinna Hance, etc. The mixed pine/broadleaf forest (MF) is a natural succession with a coniferous broadleaf ratio of about 4:6, and 70–80 years old. The main broadleaf tree species are *Schima superba Gardner & Champ.*, *Castanopsis chinensis (Spreng.) Hance*, and the coniferous species *Pinus massoniana Lamb*. The pine forest (PF) planted before 1960 belongs to the primary succession community where *Pinus massoniana Lamb* forms the only tree layer. The community composition and biodiversity are shown in Table S1.

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187 **2.2 Gross rainfall, throughfall and stemflow monitoring**

Atmospheric rainfall data was collected at Dinghushan Automatic Meteorological Station from 2001–2022. Automatic meteorological systems were used to measure atmospheric pressure (DPA501 gas-pressure meter), temperature (HMP45D sensor), relative humidity (HMP45D sensor), rainfall (SM1-1 pluviometer) and recorded with CR1000X datalogger (America Campbell). The resolution of data recording was ± 0.2 mm with a time interval of 10 min. The raw data comprised annual rainfall amounts, as well as single rainfall events with throughfall and stemflow measurements.

Throughfall and stemflow were collected in all three forest types and synchronously measured. Devices with cross-shaped collectors (1.25 m^2) attached to reservoirs (1000 L) at the bottom were used to collect throughfall. Three throughfall devices were randomly installed in each forest field (Fig. S1).

199 Half-shell plastic tubes were installed around tree trunks attached to reservoirs (1000 L) at the bottom to collect stemflow. The ratio of volume (mL) to canopy area 200 (cm²) is the stemflow (mm). A total of 24 trees were selected to measure stemflow 201 volume (Table S2). In detail, four tree species were selected in the broadleaf forest, 202 203 including Acmena acuminatissima (Blume) Merr. et Perry (SF1), Cryptocarya chinensis (Hance) Hemsl. (SF2), Gironniera subaequalis Planch. (SF3), Schima 204 superba Gardn. et Champ. (SF4), with 3 repetitions respectively. Three tree species 205 were selected in the mixed forest, including Castanea henryi (Skam) Rehd. et Wils. 206 (SF5), Schima superba Gardn. et Champ. (SF6), Pinus massoniana Lamb. (SF7), with 207 3 repetitions respectively. In the pine forest, Pinus massoniana Lamb. (SF8) was 208 209 selected as the monitoring subject with 3 repetitions.

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211 **2.3 Rainwater chemistry measurement**

For the measurement of rainwater chemistry, rainwater samples were taken in 2000,

2010 and 2022. The samples of open rainfall, throughfall and stemflow were manually 213 collected for every one-month period, respectively. The samples of open rainfall and 214 throughfall were collected with three repetitions, and stemflow with four repetitions in 215 the broadleaf forest, three repetitions in the mixed forest and three repetitions in the 216 pine forest. In total, 792 rainwater samples (108 open rainfall, 324 throughfall and 360 217 218 stemflow) were collected.

219 Rainwater samples were defrosted and filtered through 0.45 µm polypropylene membranes. Concentrations of total nitrogen (TN) and total phosphorus (TP) were 220 221 measured using an ultraviolet spectrophotometer (Lambda 25, Perkin-Elmer), and ion potassium (K⁺) was measured using an inductively coupled plasma optical emission 222 spectrometer (Optima 2000, Perkin-Elmer). The original data of TN, TP and K⁺ were 223 processed into annual flux and monthly values with the weighted average method: 224

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$$C = \frac{\Sigma c_i * V_i}{\Sigma V_i} \tag{1}$$

where C_i and V_i are the concentrations of ions (mg L⁻¹) and water sample volume (L) in 226 227 each rainfall event, respectively.

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2.4 Other measurement and statistical analysis

In the forests, plant density and canopy structure were measured every five years 230 231 since 2000. In total, 25 plots of 20 m \times 20 m (A1-A25 plots) were built on an area of 1ha to survey tree density (Fig. S1). Then, 25 plots of 5 m \times 5 m (B1-B25 plots) were 232 233 randomly set on the A1-A25 plots to survey shrub density. Finally, 25 plots of $1 \text{ m} \times 1$ m (C1-C25 plots) were randomly set on the B1-B25 plots to survey herb density. The 234 percentage of the surface area covered by plants to the total plot area is termed canopy 235 coverage (%). 25 observation plots $(1 \text{ m} \times 1 \text{ m})$ were selected in the 1 hm² area of each 236 237 forest type. LAI (Leaf area index) was measured using a LAI-2200 plant canopy analyzer with 90° view caps (Li-Cor Inc., USA). 10 observation points (distance of 238 about 10 m) were selected in the 1 hm² area of each forest type with 5 replications. 239 Growth indicators of the selected trees have been recorded: tree height (m), diameter at 240 breast height (DBH, cm), and crown area (CA, m²). Tree height was measured using a 241 laser range finder. A tape measure was used to measure the diameter of trees at a height 242 of 1.3 m, namely DBH. CA: the laser rangefinder was used to measure the maximum 243 diameter at the edge of the canopy, with multiple measurements at different points to 244 245 ensure accuracy.

The differences in throughfall and stemflow among different forests were assessed using analysis of variance (ANOVA), followed by a Tukey test for multiple comparisons between means. Mann-Kendall (MK) test was used to analyze the variation trend of annual rainfall. All statistical procedures were conducted with $\alpha =$ 0.05 threshold for significance, in the IBM SPSS statistics 22.0 software (IBM Inc.).

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252 3 Results

3.1 Rainfall and temperature characteristics

Based on the 22-year rainfall dataset from the Dinghushan area, annual gross 254 rainfall ranged between 1370.0 and 2361.1 mm (Fig. 1). 78.0% of gross rainfall 255 appeared in the wet season (April-September). The M-K test showed that rainfall was 256 significantly decreasing trend from 2001–2007 (UF < 0, P < 0.05), and shifted into a 257 significantly increasing pattern from 2012–2022 (UF > 0, P < 0.05). Moreover, 2008 258 and 2011 (the intersection of UF and UB) were the mutation time steps of the rainfall 259 trend (P < 0.05). Anomalies were revealed in the temporal variability (coefficient of 260 variation, CV of 16.6%) in annual rainfall (Fig. 1a). Anomalies varied at -426.4-476.8 261 mm and -258.0-471.4 mm in the wet season and dry season, respectively. By 262 comparison, the dry season experienced greater variation with CV of 40.4% than the 263 wet season with a CV of 21.7%. Besides, annual rain days tended to decrease over time 264 from 2012 to 2021(Fig. 1b). Based on five rainfall classifications, it was shown among 265 22 years that rainfall <10 mm accounts for about 68.5% of total rain days (2856), while 266 rainfall >50 mm account for about 4.9%. Besides, annual temperature changed between 267 21.8 °C and 23.3 °C over 22 years. The M-K test showed a statistically significant 268 increase in temperature for 8 years out of 22 years (UF > 0, P < 0.05). Moreover, 2005 269 and 2013–2014 were the mutation time steps of the temperature trend (P < 0.05). 270

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272 **3.2 Variability of throughfall**

Rainfall redistribution (throughfall and stemflow) among the three forests all experienced differing magnitudes for 22 years (Fig. 2). Annual throughfall was concentrated between 954.2 mm and 2192.6 mm. The M-K test showed that throughfall was in a significantly decreasing trend at first (UF < 0, P < 0.05) and then shifted into a significant increase (UF > 0, P < 0.05) from 2001–2022, similar to the trend of open rainfall. Differently, a mutation time step of throughfall occurred in 2008, 2011 and

2021 in the broadleaf forest, 2008 and 2011 in the mixed forest, 2006, 2008, 2011, and 279 2021 in the pine forest (P < 0.05). The throughfall ratio varied significantly both at 280 event and interannual scales (Fig. 3a, b and c). The median annual throughfall ratio in 281 the broadleaf forest varied between 60% and 120% with a CV of 16.4% from 2001-282 2022. The median throughfall ratio in the mixed pine and broadleaf forest varied 283 between 80% and 110% with a CV of 9.7%. The median throughfall ratio in the pine 284 forest varied between 59% and 110% with a CV of 11.8%. Therefore, the throughfall 285 ratio was characterized by a low variability over an annual-time scale (CV < 20%). 286 287 Besides, some differences in throughfall ratio were found among the three forest types based on rainfall classifications (Fig. 3d). For rainfall events <10 mm, the throughfall 288 ratio range in the broadleaf forest was 30%–70%, while in the other two forest types, it 289 was 15%–85%. The mean value of the throughfall ratio was small in the broadleaf forest 290 (53.9%), though no significant differences among the three forests (P > 0.05) were 291 detected. For rainfall events <50 mm, no significant difference in throughfall ratio 292 among the three forest types was found (P > 0.05). However, the median values of the 293 throughfall ratio in the pine forest (90.0%) and the mixed forest (89.4%) were both 294 significantly larger than that in the broadleaf forest (83.7%) for rainfall events >50 mm. 295

296 CV values of throughfall based on all the rainfall event classifications were drawn in the Fig. 4a. Results showed that the median CV of throughfall in the pine forest 297 (15.2%) was lower than that for the broadleaf forest (21.7%) and for the mixed forest 298 (26.3%) for rainfall events <10 mm. For rainfall events >10 mm, small differences in 299 median CV among the three forest types were shown. Meanwhile, CV values decreased 300 with the increasing rainfall events, eventually falling to 3.5%–4.3%. Besides, CV values 301 302 of throughfall based on the interannual scale were drawn in the Fig. 5a, b and c. Annual CV values among different forest types showed different fluctuations over time. The 303 304 medians of CV in annual, wet and dry seasons presented different orders in different years. According to linear fitting, significant negative correlations were found in the 305 median of CV_{TF} in the mixed forest over time (r = 0.63, P < 0.01). In addition, the fitted 306 result of in total 740 rainfall events in 22 years showed that CV values of throughfall 307 significantly decreased with increasing gross rainfall (Fig. S2). 308

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310 **3.3 Variability of stemflow**

Annual stemflow was concentrated between 9.0 and 119.7 mm over 22 years (Fig.2). More stemflow was collected in the broadleaf forest and mixed forest than in the

pine forest. The M-K test showed that stemflow of both broadleaf forest and mixed 313 forest was decreasing at first (UF < 0, P < 0.05) and then shifted into an increase (UF > 314 0, P < 0.05), different from the continuously increasing trend of pine forest (UF > 0). 315 The mutation time steps of stemflow occurred in 2008 in the broadleaf forest, 2011 in 316 the mixed forest, 2006, 2012 and 2015 in the pine forest (P < 0.05). Stemflow ratio 317 changed significantly both at the event and interannual scales (Fig. 3a, b and c). For 22 318 years, the median annual stemflow ratio in the broadleaf forest varied between 1.3% 319 and 5.4% with a CV of 56.2%. The stemflow ratio of mixed forest varied between 1.5% 320 321 and 4.4% with a CV of 38.6%. In the pine forest, it varied between 0.3% and 1% with a CV of 50.9%. This indicated that the stemflow ratio was characterized by an extremely 322 high variability over time. Similar to the seasonal throughfall ratio, the medians of 323 stemflow ratios in annual, wet and dry seasons presented different orders in different 324 years. Besides, the stemflow ratio significantly changed among tree species and rainfall 325 classifications (Fig. 3e). By comparison, the stemflow ratios of the SF1 and SF2 trees 326 in the broadleaf forest were both higher in all the tree species for the rainfall events <50 327 328 mm. However, for strong events (>50 mm), the stemflow ratio of the SF5 tree in the mixed forest was highest for all tree species, followed by the trees in the broadleaf forest. 329 330 For all the rainfall events, the stemflow ratio of SF7 in the mixed forest and SF8 in the pine forest were both lower than that for other tree species. 331

CV values of stemflow based on rainfall event classifications were drawn in Fig. 332 4b. By comparison, stemflow varied more than throughfall across all rainfall events, 333 with CV_{SF} values of 25%–130%. Median CV of stemflow in the pine forest was always 334 lower (45%–68%) than that for the other two forest types (56%–120%). CV values of 335 stemflow based on an interannual scale changed over time among different forest types 336 (Fig. 5d, e and f). The medians of CV_{SF} in annual, wet and dry seasons presented 337 different orders in different years. By comparison, CV_{SF} was always greater than CV_{TF} , 338 and the interannual fluctuation of CV_{SF} was also stronger than CV_{TF}. According to linear 339 fitting, significant negative correlations were found in the median of CV_{SF} in the 340 broadleaf forest over time (r = 0.73, P < 0.001)). In addition, the fitted result of in total 341 740 rainfall events in 22 years showed that CV values of stemflow both significantly 342 decreased with increasing gross rainfall (Fig. S2). 343

345 **3.4 Rainwater chemistry**

Rainwater (open rainfall, throughfall and stemflow) chemical properties (TN, TP 346 and K^+ concentration) were measured in the three forest types, respectively. All of TN, 347 TP and K^+ values are presented in the order stemflow > throughfall > open rainfall (Fig. 348 6a, b and c). However, changes of TN, TP and K^+ were different for the three forest 349 types in 2000, 2010, and 2022. For instance, in 2000 and 2010, TN values of throughfall 350 and stemflow decreased for both from pine forest to mixed forest to broadleaf forest, 351 while no such result could be confirmed in 2022. Similarly, TP values of throughfall in 352 353 the broadleaf forest were 1.3 times higher than in the pine forest in 2022, while TP values in the pine forest were 6.8 times higher than that in the broadleaf forest in 2000. 354 K^+ values of stemflow in 2010 (6.76 mg L⁻¹) and 2022 (6.22 mg L⁻¹) were higher for 355 the broadleaf forest than for the pine forest (3.76 mg L^{-1} and 2.46 mg L^{-1}), which was 356 different from that in 2022. 357

TN, TP and K⁺ fluxes of stemflow were < 10 kg ha⁻¹ a⁻¹, 0.2 kg ha⁻¹ a⁻¹, 6 kg ha⁻¹ 358 a⁻¹, respectively, and all lower than those of throughfall and open rainfall (Fig. 7d, e and 359 f). In 2000, 2010, and 2022, TN fluxes (39.4–87.4 kg ha⁻¹ a⁻¹) were 1.2–1.8 times higher 360 than that of open rainfall, 3.3–28.0 times higher than that of stemflow. TP fluxes (1.1– 361 2.7 kg ha⁻¹ a⁻¹) were 1.0–2.3 times higher than that of open rainfall, 8.7-31.4 times 362 higher than that of stemflow. K^+ flux (21.5–59.2 kg ha⁻¹ a⁻¹) was 2.2–8.1 times higher 363 than that of open rainfall, 2.2–26.8 times greater than that of stemflow. In addition, TN, 364 TP and K⁺ fluxes of stemflow increased with succession from primary to climax, 365 namely pine forest<mixed forest
broadleaf forest. Different from this, differences in 366 chemical fluxes of throughfall were not found among different forests, nor among 367 different periods. 368

Moreover, monthly chemical concentrations in rainfall, throughfall and stemflow 369 showed distinct changes (Fig. 7). Monthly TN, TP and K⁺ concentrations of rainfall 370 were always lower than those of stemflow for all trees. Monthly TN, TP and K^+ of 371 stemflow in the dry season were generally higher than in the wet season. High monthly 372 TN concentrations of stemflow with SF6 of mixed forest and SF8 of pine forest were 373 found, especially in the dry season with maximum TN concentrations of 27.59 mg L⁻¹ 374 at SF6 and 19.94 mg L⁻¹ at SF8, respectively. Differently, a high monthly K⁺ 375 concentration of stemflow at SF4 in broadleaf forest was found, with a maximum K⁺ 376 concentration of 25.17 mg L^{-1} in the dry season. 377

379 4 Discussion

4.1 Open rainfall partitioned to throughfall and stemflow

Studies in forests have confirmed that throughfall volume increased with 381 increasing gross rainfall at the event scale, accounting for 60%-80% of gross rainfall 382 (Ji et al., 2023; André et al., 2011; Carlyle-Moses, 2004). Throughfall ratio changed 383 384 over time and showed different fluctuations among different forests (Fig. 3). During light rainfall events with rainfall amounts <10 mm, a low proportion of raindrops would 385 reach the ground as throughfall, as the tree canopy intercepts almost all the incoming 386 raindrops. Specifically, high canopy coverage in broadleaf forests can reinforce 387 388 raindrop intercept (Brasil et al., 2018; Ponette-González et al., 2010), consequently generating a lower throughfall ratio than those in the mixed forest and pine forest (Fig. 389 3). During moderate rainfall events (10–50 mm), given that the intercept effect of the 390 wetting tree canopy was weakened (Shinohara et al., 2015), throughfall ratio was in a 391 high and steady state. As the gross rainfall increases further (>50 mm), significant 392 differences of throughfall ratio were found among the three forests. The throughfall 393 ratio was significantly lower in the broadleaf forest than those in the other two forests. 394 Likewise, such differences due to the rainfall event class also appeared in other forest 395 studies with stands formed by beech, pine in monocultures and mixed pine-beech 396 397 (Blume et al., 2022). Influenced by forest stand characteristics, throughfall therefore indicated different forest water budgets. 398

Stemflow of forests was variably controlled by tree species, on average accounting 399 for about <10% of gross rainfall, and partly even lower (<1%) (Sun et al., 2018; André 400 et al., 2008; Crockford and Richardson, 1990). In our study site, the lowest stemflow 401 (<1%) was collected in the pine forest, though weakly increasing with rainfall 402 classifications (Fig. 3). Stemflow ratio in the broadleaf forest was maintained at 5%-403 10% without the effect of rainfall amount seemingly. In detail, stemflow ratio of pine 404 forest (SF8) was significantly lower than those of broadleaf forest (SF1~4). In the 405 mixed forest, broad-leaved trees (SF5 and SF6) have larger stemflow than pine trees 406 407 (SF7). However, for some rainfall events, a particularly low proportion of stemflow in the broadleaf forest and an extraordinarily high proportion in the pine forest were 408 caught. This implied the key role of rainfall conditions (e.g., intensity, duration) and 409 410 tree species with tree traits (e.g., branch angle), consistent with reported studies e.g., in an evergreen forest (Chen et al., 2019; Bruijnzeel et al., 2011) and pine forest (Pinos et al., 2021; Crockford and Richardson, 1990). Moreover, ANOVA showed significant differences in stemflow ratio among tree species and rainfall classifications (P < 0.001) (Table 1). This indicated that rainfall and tree species simultaneously affect stemflow. Branch inclination angle, canopy cover, tree height and DBH of tree species proved to be key factors in stemflow yield (Levia et al., 2015).

417 Throughfall and stemflow were generally enriched in chemical concentration compared with open rainfall due to leachable canopy/stem ion pools (Jiang et al., 2021; 418 419 Van Stan et al., 2017; Zimmermann et al., 2007). In our study, the concentration of K⁺ in stemflow was 16 times higher than that in open rainfall and in throughfall reached 420 up to 11 times higher than open rainfall (Fig. 6). Similar results were also found in 421 forestry plantations (Acacia mangium and Dimocarpus longan) of South China (Shen 422 et al., 2013), or in Oriental beech (Fagus orientalis Lipsky) trees in Northern Iran 423 (Moslehi et al., 2019), indicating strong K⁺ leaching from the canopy. Even so, 424 throughfall was generally characterized by high fluxes compared to open rainfall 425 426 followed by stemflow, it thus is the largest contributor to wet deposition. Meanwhile, the TN flux of throughfall was greatest in the pine forest in 2010, the TP flux of 427 428 throughfall was greatest in the broadleaf forest in 2000, and the K⁺ flux of throughfall was greatest in the mixed forest in 2010. It should be noted that the differences in 429 430 rainwater chemistry shifted over time among the three forests. Accordingly, throughfall and stemflow via the canopy and stem to the soil are a significant contributor, and their 431 432 long-term effect on ecosystems needs more scientific attention (Fan et al., 2021). After all, atmospheric wet deposition provides important nutrient amounts for ecosystems, 433 but also imposes a considerable burden on the ecosystems in general. For instance, N 434 enrichment and P limitation have proven to have different effects on soil carbon 435 sequestration, microbial community composition and forest productivity, especially in 436 tropical and subtropical forest ecosystems with highly weathered soils (Zheng et al., 437 2022; Li et al., 2016; Huang et al., 2012). Besides, throughfall and stemflow was mainly 438 characterized by low chemical concentrations in the wet season and high concentrations 439 440 in the dry season. Primary reasons for seasonal rainwater chemistry may be attributable to moisture source associated with frontal weather systems and gradually depleting 441 effect with increasing rainfall amount (Dunkerley, 2014; Germer et al., 2007). The 442 present study in subtropical forests and previous studies in tropical forests and European 443 temperate forests all exhibited variable rainwater chemistry in throughfall and stemflow, 444

both spatially and temporally (Zimmermann et al., 2007; Staelens et al., 2006; Seiler
and Matzner, 1995). The chemical concentration of rainfall redistribution was also
affected profoundly by canopy and stem parameters of tree species (Tonello et al.,
2021b; Chen et al., 2019). In our study, some differences in TN, TP and K⁺ were also
found among SF1~SF8 due to tree-species-specific effects (Legout et al., 2016; De
Schrijver et al., 2007).

451

452 **4.2 Long-term changes in rainfall in forests**

453 Rainfall regimes induce divergent spatially hydrological changes (Wu et al., 2024). Likewise, our study found that the throughfall of forests experienced a decrease 454 followed by an increase from 2001-2022, similar to the trend of open rainfall, and 455 stemflow showed characteristic trends in different forests especially in the pine forest 456 (Fig. 1). This suggested that the complexity of forest structure and rainfall amount and 457 their change exacerbated the spatio-temporal variability of throughfall and stemflow. 458 Firstly, interannual variability of forest structure (e.g., canopy coverage, leaf area index) 459 and tree parameters (e.g., height, DBH, and CA) made throughfall and stemflow 460 distribution uncertain (Yue et al., 2021). From 2001 to 2022, changes in forest structure 461 462 were confirmed in all three forests, such as changes in plant density, canopy coverage and LAI (Fig. 8). Throughfall ratio and stemflow ratio in the succession forest systems 463 all varied over time accordingly. Similarly, driven by forest structure (e.g., tree density, 464 species dominance), a six-year dataset from the Brazilian Atlantic Forest showed that 465 the spatial variability of throughfall over time was less stable (Rodrigues et al., 2022). 466 Besides, the variation of stemflow (CV_{SF}) was larger than that of throughfall (CV_{TF}) 467 (Fig. 4), which probably was attributed to the differences of tree species in stemflow 468 (Fig. 3). For a forest succession, a 17 years' study showed that the shift from 469 470 monoculture Japanese red pine to mixture of red pine, evergreen oak and deciduous trees made stemflow significantly increasing (Iida et al., 2005). Likewise, for the forest 471 succession in Dinghushan area, stemflow ratio in the broadleaf forest and mixed forest 472 were both higher than that in the tree-monospecific pine forest. High plant density (tree 473 and shrub) and LAI in broadleaf forest and mixed forest conduce to rainwater 474 interception of multi-canopy trees through more leaves and angled branches, which 475 potentially enhanced stemflow (Fig. 8). Indeed, some differences of rainfall 476 redistribution appeared in multi-layered vegetative structure. An experiment on 477 vegetation communities with a complex multi-layered structure found that interception 478

479 loss from shrubs was two times higher than from trees, and smaller trees generated stemflow more efficiently than the higher ones (Exler and Moore, 2022). Based on the 480 22 years' data from the forest community survey in our study site, forest canopy 481 parameters (e.g., coverage and LAI) of trees and shrubs showed variation over time 482 from 2001 to 2022 (Fig. 8). In the broadleaf forest, plant density of trees and canopy 483 coverage of shrubs showed a slight increment compared to the other two forest types, 484 though LAI was decreasing. During this period, interannual throughfall ratio and 485 stemflow ratio showed significant change over time (Fig. 3), implying the role of 486 487 interannual variation of forest structure in the rainfall redistribution process.

Secondly, ongoing rainfall changes with different magnitudes favor the different 488 levels of rainfall redistribution over time (Lian et al., 2022). At the event scale, 489 throughfall and stemflow proportions of forests were both low with rainfall events <10 490 mm. The variations of throughfall and stemflow were both larger for gross rainfall <10 491 mm than events >10 mm. Rainfall threshold associated with the canopy interception 492 capacity had an impact on throughfall and stemflow generation (Zabret et al., 2018; 493 André et al., 2008; Durocher, 1990). After the raindrop capacity of the canopy reached 494 its peak, throughfall and stemflow were documented to match the gross rainfall. 495 496 Therefore, low proportions and high spatial variability appeared before the rainfall threshold, and after that, relatively high proportions and low variability until a stable 497 level were observed in the three forests. Moreover, at the interannual scale, the rain days 498 in different magnitudes presented fluctuation over 22 years (Fig. 1). This fluctuation of 499 500 rain days and its magnitude distribution potentially regulated the long-term changes of open rainfall partitioned to interception loss, throughfall and stemflow. Consequently, 501 502 throughfall and stemflow, influenced by the comprehensive effect of rainfall regimes and forest structures, presented spatiotemporal variability at different levels (Fig. 3-6). 503 504 From a long-term perspective, changes in rainfall redistribution potentially make forest water and biogeochemistry budgets more complex. Further knowledge of the long-term 505 accumulative effect of rainfall redistribution on forest water and chemistry (e.g., soil 506 and plant) is needed in the future. 507

508 Throughfall and stemflow are part of rainfall and are key players in the water cycle 509 process. Based on the connection of the water cycle to precipitation and temperature 510 and under the background of climate change, the frequency of extreme events (heavy 511 rainfall, droughts) needs to be anticipated in the effect on rainfall redistribution and 512 solute transport within forests, which in turn may affect the water cycle and

biogeochemical cycles (Blume et al., 2022). In this study, it should be noted that the 513 2008 rainfall data can be used as an example of an extreme event. In 2008, extreme 514 weather events occurred in South China. Freezing events occurred in the dry season, 515 and continuous heavy rain and typhoon events occurred in the wet season. Gross rainfall 516 was larger than in other years, with an annual rainfall of 2361.1 mm (22-year average 517 518 annual rainfall of 1848.6 mm) (Fig. 1). At the same time, a total of 26 throughfall events 519 were collected in 2008. The throughfall and stemflow trend of different forests presented different degrees of disturbance under the background of mutation of open 520 521 rainfall (Fig. 2). In this process, the driving effect of forest structure and rainfall on throughfall and stemflow mutation is synchronous. More data and modeling are needed 522 to support the relevant study about the effect of climate change on rainfall redistribution 523 in the future. 524

525

526 **5 Conclusions**

The current study investigated long-term changing characteristics of rainfall 527 redistribution along a subtropical forest succession sequence with pine forest (PF), 528 mixed pine and broadleaf forest (MF), and monsoon evergreen broadleaf forest (BF). 529 530 Firstly, in the 740 measured rainfall events, the throughfall ratio changed from BF < MF < PF, and the stemflow ratio changed from BF > MF > PF. The variation of 531 stemflow was higher (CV > 50%) than that of throughfall (CV < 25%). Secondly, 532 throughfall and stemflow of the investigated forests experienced a decrease followed 533 534 by an increase from 2001–2022 (except stemflow of the pine forest), similar to the trend of open rainfall. Driven by rainfall and forest factors, the interannual variability of both 535 throughfall and stemflow in the broadleaf forest was higher than those in the mixed 536 forest and pine forest, which was different from that of annual open rainfall. 537

538 For rainwater chemistry, differences in the element fluxes in throughfall and stemflow among the three forest types were confirmed based on data from 2001, 2010, 539 and 2022. On average, TN and TP fluxes of throughfall developed from BF < MF < PF, 540 while K^+ flux of throughfall changed from BF > MF > PF. Over time, rainwater 541 542 chemical concentrations were lower in the wet season than in the dry season. Given the smaller proportion of open rainfall, stemflow chemical fluxes varied less among forest 543 types and over time, though tree species exactly represented the differences in stemflow 544 chemistry. Nevertheless, its funnel effect on soil and plants over time still deserves more 545 scientific attention in the future. 546

547	The above results indicate that the water volume and chemistry of rainfall during
548	redistribution processes under forests represent not the same trend as open rainfall over
549	time, and throughfall and stemflow depend on the effect of rainfall and forest factors.
550	This study thus provides insight into the rainfall redistribution process by linking the
551	long-term change of rainfall patterns with a subtropical forest succession sequence.
552	
553	
554	Code and data availability. The data used to derive to the conclusions of the present
555	study are freely accessible. All the data were obtained from the CNERN dataset
556	(http://dhf.cern.ac.cn/meta/metaData).
557	
558	Author contributions. WJZ: conceptualization, investigation, data analysis, writing,
559	visualization. TS and SS: reviewing, supervision. QMZ and GWC: resources, data
560	curation, LHW: reviewing, JXL and XX: reviewing, funding acquisition, supervision
561	
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582

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Tables 842

843

844 Table 1. Correlations between throughfall and stemflow and rainfall and forest factors

	Gross rainfall	DBH	CA	Height	LAI	
Throughfall	0.72***				-0.58**	
stemflow	0.77***	0.65*	-0.75**	0.54**	-0.55**	
DBH: diameter at breast height; CA: crown area; LAI: leaf area index. * $P < 0.05$, ** $P < 0.01$, *** $P < 0.$						
0.001						
Table 2. Analysis of variance (ANOVA) for throughfall and stemflow affected by rainfall						
classifications and tree species						
Summary of Al	NOVA T	hroughfall			Stemflow	
Rainfall classifi	ication (R) <	0.05	Rainfall o	classification (R)	< 0.001	
Forest type (F)	<	0.001	Tree spec	cies (T)	< 0.001	
$\mathbf{R} \times \mathbf{F}$	0	.861	$\mathbf{R} \times \mathbf{T}$		< 0.001	

Forest type (F) < 0.001 $\mathbf{R} \times \mathbf{F}$ 0.861

 $\alpha = 0.05$ 855



Figure 1. (a) and (b) rainfall and temperature in Dinghushan Biosphere Reserve in Southern China from 2001–2022, (c) and (d) annual rainfall and temperature, (e) and (f) rainfall and temperature statistic of Mann-Kendall test, respectively. (g) Anomaly of annual rainfall from 2001–2022, (h) annual raining days in five classifications. UF (Unadjusted Forward) > 0 indicate a continuous increasing trend (P < 0.05). The intersection points of UF and UB (Unadjusted Backward) is the mutation time point. Within the confidence interval [-1.96, 1.96], the variable presents a significantly mutation growth state at this time point (P < 0.05).



868Figure 2. (a) and (b) Annual throughfall and stemflow in the broadleaf forest (BF), mixed pine and869broadleaf forest (MF) and pine forest (PF) from 2001–2022, respectively, (c) ~ (h) rainfall and870stemflow statistic of Mann-Kendall test, respectively. UF (Unadjusted Forward) > 0 indicate a871continuous increasing trend (P < 0.05). The intersection points of UF and UB (Unadjusted872Backward) is the mutation time point. Within the confidence interval [-1.96, 1.96], the variable873presents a significantly mutation growth state at this time point (P < 0.05).



881 Schima superba Gardn. et Champ. (SF6), Pinus massoniana Lamb. (SF7); pine forest: Pinu 882 massoniana Lamb. (SF8). Different letters indicate a significant difference at P < 0.05



Figure 4. Box plots of coefficient of variation (*CV*, %) in (a) throughfall (TF) and (b) stemflow (SF)
in broadleaf forest (BF), mixed pine and broadleaf forest (MF) and pine forest (PF) based on
the rainfall classifications. Different letters indicate a significant difference at *P* < 0.05

Figure 5. Box plots of coefficient of variation (*CV*, %) in (a, b, and c) throughfall (TF) and (d, e, and f) stemflow (SF) in the three forests from 2001 to 2022. Medians of annual *CV* were fitted.

r: Pearson coefficient of correlation; *: P < 0.05, **: P < 0.01, ***: P < 0.001

Figure 6. Concentrations and fluxes of TN, TP and K⁺ of gross rainfall (GR), throughfall (TF) and
 stemflow (SF) in the broadleaf forest (BF), mixed forest (MF) and pine forest (PF) in 2000,

896 2010 and 2022, respectively.

Kigure 7. Monthly concentrations of (a) TN, (b) TP and (c) K⁺ of throughfall (TF1) and stemflow (SF1, SF2, SF3 and SF4) in the broadleaf forest, throughfall (TF2) and stemflow (SF5, SF6 and SF7) in the mixed forest, throughfall (TF3) and stemflow (SF8) in the pine forest.

